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ChE Preliminary Exam Outline

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**Agenda for peep meeting**

* Yay NSF! Kinda yay Ford! Yay Point!
  + Any tips for funding interviews? (Point)
* Align on iteration timeline
* Discuss/align on prelim outline

**Topics for prelim discussion**

* Confirm some assumptions I’m working off of:
  + This is not a contract, and I can take my thesis in another direction if something else interesting piques my interest in a year.
  + The proposed work in the prelim should be somewhere between boring and ambitious, rather than really go out on a risky limb.
* Feedback requested:
  + How specific am I expected to be about “distant” plans? (Seems to really vary by whose prelim I’m looking at)
  + Am I still thinking too big? Any advice on being granular on such a long time scale?

**Structure**

1. Working title
2. Intro/Motivation
3. Background
4. Proposed work: Understand how particle anisotropy (specifically shape) impacts self-assembly in drive non-equilibrium systems
   1. Aim 1: Predict structures and formation kinetics of self-assembled anisotropic hard particle systems (2D, then expand to 3D?)
   2. Aim 2: Use activity to direct self-assembly and stabilize steady state structures in otherwise-passive systems (3D)
   3. Aim 3: Use activity to induce reconfigurability in material systems (3D)
5. Timeline
6. Reference: Other cool ideas to draw from

**Working title**: [Studying non-equilibrium self-assembly in active, driven systems]

**Intro/Motivation**:

* **Reconfigurable systems** hold great promise for enabling adaptive material applications such as next-generation self-healing materials, wet computing information storage, and responsive photonic devices
  + Existing examples of reconfigurable materials are limited to macroscale systems for printed or otherwise manually arranged particles
  + However, such methods are infeasible at the nanoscopic scales required for realization of the aforementioned applications
* **Equilibrium self-assembly** can be used to nanofabricate ordered structures by allowing atoms, molecules, or colloidal particles to sample many potential configurations through thermal fluctuations
  + The resulting self-assembled structure is the configuration that minimizes the free energy
  + In equilibrium systems, colloidal-scale self-assembly is a well-characterized process
  + Particle anisotropy, specifically shape, has been shown to determine the structure of self-assembled colloidal crystals, even in the absence of explicit inter-particle interactions
* **Experimentally**, many **synthetic colloidal systems** do not assemble reliably or robustly
* In contrast, **biologically assembled materials** often reliably assemble into metastable, non-equilibrium states that allow them to rapidly respond to external stimuli structurally and functionally
  + Many of these biological structures are stabilized by the constant input of energy from ATP, which provides the driving force for non-equilibrium self-assembly in these systems
* This robust self-assembly inspires engineers looking to design synthetic nanofabrication processes
  + However, the fundamental physics of such driven non-equilibrium self-assembly remains poorly understood
* By understanding the principles governing driven non-equilibrium self-assembly, we can eventually design pathways to robustly self-assembly reconfigurable, adaptive materials for nanoscale applications

**Background**:

* “Active matter” describes driven non-equilibrium systems in which a self-propulsive driving force is applied to each particle
  + These systems display a wealth of non-equilibrium behavior, such as a collective motion (e.g. swarming) and giant density fluctuations (e.g. clustering)
* Experimental and theoretical active matter research has focused primarily on the behavior and self-assembly of isotropic particles
  + While useful for defining the basic physics of these systems, such particles do not have a natural “alignment rule” allowing for torques to be effected on neighboring particles
  + However, anisotropic particles with shape *are* able to alter the orientations of neighboring particles, and we know that particle shape alone is sufficient to direct self-assembly in equilibrium systems
* Thus, a natural question arises: how does particle anisotropy, specifically shape, impact self-assembly in drive non-equilibrium systems?

***(Section on proposed work begins on next page)***

**Proposed work**: **Understand how particle anisotropy (specifically shape) impacts self-assembly in driven non-equilibrium systems**

* Computational capabilities allow the study of active systems on large enough time and length scales to be predictive
* Experimentally determining the key drivers of self-assembly is prohibitively time-consuming
* **Proposal**: Computationally develop an understanding of the drivers of non-equilibrium self-assembly to be able to guide experimental work towards systems of interest

**Timeline:**

I’ll put together a nice Gantt chart that ties all this together.

***Aims spelled out on the following pages.***

**1) AIM #1**: **Predict structures and formation kinetics of self-assembled anisotropic hard particle systems (2D, then expand to 3D?)**

**Output:**

a) Collective motion onset versus shape (n-gons), driving force, density

b) Cluster structure versus equilibrium crystal structure

c) Theory for kinetics of collective motion that incorporates shape

d) *Anything needed beyond this kinetic theory to claim “prediction”?*

e) *I will probably need to expand this to 3D for aims #2 and #3*

**Hypothesis and rationale for it:**

* Self-assembled structures in driven systems can be related to the system density, applied driving force, and anisotropy of the particles (cite isotropic systems)

**Key preliminary results**:

*In 2D:*

* Cluster onset diagrams for n-gons 4-8(+?)
* Cluster structure for n-gons 4-8(+?)
* (Maybe) Comparison with equilibrium structure for n-gons 4-8(+?)
* (Maybe) Isaac/my collision theory for general active particles, with some work on how it might be extended to take shape into account

**Experimental design/specific methods**:

* *Methods*: Active particles will be modeled using HOOMD-blue molecular dynamics, and open source software package built to run in parallel on GPUs and CPUs, and developed and maintained by the Glotzer lab. Anisotropic shape interactions are accounted for with a discrete element method, borrowed from the granular matter community (read up on this) and implemented in HOOMD-blue.
* *Analysis*: Clusters of active particles have previously been shown to assemble into “active crystals”. The emergence of clusters versus driving force and density can be compared with the sparse literature on anisotropic active particles, while structure can be checked against known equilibrium assemblies. Equilibrium self-assembly of anisotropic particles can be validated using hard-particle Monte Carlo in HOOMD-blue, by equating active driving force to system pressure.

**Complications/alternative strategies**: These Brownian simulations take a long time to run (out-standing issue)

**2) AIM #2: Use activity to direct and stabilize self-assembly in passive systems (probably need to go to 3D)**

**Output**

a) TBD

**Hypothesis and rationale for it**:

* (I’m thinking of some of the work out of the Djikstra group mixing active and passive particles in assembly)

**Key preliminary results**: N/A

**Experimental design/specific methods**:

* Choose systems that can get easily kinetically trapped en route to their true equilibrium structure (i.e. have well-known metastable states)
  + Will need to do some talking to folks in lab to figure out what good candidate systems might be, this is not something I’ve actively looked for before
* Add in varying ratios of passive/active particles (same shape) and varying driving forces, develop phase diagram of final structure formed
  + Possible to make a unifying theory of how the driving force corresponds to the free energy difference between the metastable and final states of the system?
* Repeat with binary mixtures; what happens if active shape and passive shapes have incommensurate final structures?

**Complications/alternative strategies**: These MD simulations may be very computationally expensive

**3) AIM #3: Use activity to induce reconfigurability in material systems (probably need to go to 3D)**

**Output**

a) TBD

**Hypothesis and rationale for it**:

* If aim #2 is achievable, then we’ve shown that we can use active particles to direct self assembly; can we then design a system that has multiple “states” in it, depending upon the driving force applied to the active particles?
* *Importance*: If reconfigurability in non-equilibrium allows us to reach otherwise inaccessible structures (Aim #2), this would be a promising nanofabrication approach. Additionally, reconfigurability-induced switching of a material’s structure could be explored as a way of storing information (“memory”) in a material.

**Key preliminary results**: N/A

**Experimental design/specific methods**:

* TBD

**Complications/alternative strategies**: TBD

**REFERENCE: Other cool project ideas**

*These either don’t fit the story in this prelim so well, or seem too “out there” to talk about during a prelim exam. Some could probably be re-worked into a decent prelim section.*

Other active matter ideas with shape and non-equilibrium self-assembly

*Some low-hanging, some not*

* Extend all this to 3D
* Evaluate the impact of hydrodynamics on active anisotropic self-assembly
* Change shape throughout a simulation—does the activity stabilize the original structure, or does cluster structure change?
* Work to develop unifying “thermodynamic” theory for active matter (though this is something more in Baskaran’s, Brady’s wheelhouse)
* In a dense system of passive particles, can we use active particles to change the self-assembled structure? How many do we need to have an effect? Is there a critical volume fraction of one type of particle that will lead to this transition?
* Are there self-assembled structures that are only stable at higher pressure (probably have to go to 3D for this), and can we use some number of active particles in the system to affect an active pressure that can stabilize the system?

Use kirigami self-assembly as a means of studying structural entropy

* Can we define nets that have more “information” than others?
* Can we define pathways that have more “information” than others?

Novel mechanisms

* In systems of shapes, could we use ratcheting/local symmetry breaking leading to a global driving force?
  + Alternately, see if shape could be used to convert a translational driving force into primarily rotational motion
* Take driving force examples from the cell into our synthetic systems:
  + ATP acts by coupling with reactions to make unfavorable reactions favorable; is there a way we could incorporate that into having particles with favorable/unfavorable interactions
  + In Na/K pumps, ATP phosphorylates them into an unfavorable configuration, which which they resolve by reconfiguring in some way and pumping Na/K in the process

Biomimicry

* Minimum sufficient cell model
* Model of durotaxis?

Colloidal robots

* “Terminator vicsek model”
* Designing behavior into a particle with shape
* Patchy colloidal robots ala the MIT sand robots, build up a driving force so that across the system appears randomly distributed